

In the United States Patent and Trademark Office

Applicant : Yoo et al.  
Serial No. : 09/581,804 Examiner : R. Lee  
Filed : June 16, 2000 Group Art Unit : 1713  
For : THERMOPLASTIC TRANSPARENT RESIN COMPOSITION AND  
METHOD OF MANUFACTURING SAME

DECLARATION UNDER 37 C.F.R. Section 1.132

I, Lee ChanHong, declare and say:

That I am a citizen of Korea and I reside at L Ghana Apt 102-901, Shinseong-dong, Yuseong-gu, Daejeon, Korea.

That I am one of the coinventors in the above-identified patent application;

That I graduated in 1978 from Univ. of Seoul located in Seoul, Korea with a Bachelor Degree in Chemical Engineering. I graduated in 1988 from Univ. of Oklahoma located in Norman, Oklahoma, USA with a Doctorate Degree in Chemical Engineering;

That since 1988, I have been working in the field of Emulsion Polymerization. I have been employed by LG Chemical Ltd. since 1988, and part of this time has been spent in this field. Since 1997, I have been developing Transparent ABS Resin in the ABS Resin Development Department of LG Chemical Ltd.

That I have been granted 23 patents on emulsion polymerization and I am the author of 4 papers published in academic journals in the emulsion polymerization technology;

That I am familiar with the above-identified United States patent application Serial No. 09/581,804 and with the following references cited by the Examiner:

U.S. Patent No. 4,767,833 and JP 56-41216;

That Claims 1 and 8 (Claim 1 was amended to be limited to Claims 3 and 4) have an inventive step for the reasons set forth below:

(1) Through the tests below (Reference Examples 1 and 2, and Example 1), I found that the gel content and the swelling index of rubber greatly influence transparency and shock resistance of thermoplastic transparent resin. I described this fact on page 5, lines 13-17 of the specification

of the present application and defined the technical aspect of limiting the gel content and the swelling index in Claims 3 and 4 of the specification as originally filed. Even though the specification does not describe the tests below, the tests were conducted at the time of the completion of the present invention. Further, the test results are described in the specification and claims as mentioned above.

The tests below support the fact that the gel content and the swelling index of rubber should be adjusted in order to provide a thermoplastic transparent resin, which has excellent transparency, latex stability and shock resistance.

[Reference Example 1]

The same method as in Example 1 of the specification of the present application was used to manufacture a small aperture rubber latex, but (1) 0.6 parts by weight of a molecular weight controlling agent (TDDM) was added instead of 0.3 parts by weight of the same, (2) 0.15 parts by weight of TDDM (tertiary dodecyl mercaptan) was added again instead of 0.05 parts by weight of the same, and (3) the reaction temperature was decreased from 65 °C to 60 °C. A large aperture rubber latex was manufactured by use of the obtained small aperture rubber latex, through the same method as that of Example 1. Graft copolymerization material was manufactured using the same method as that of Example 1.

[Reference Example 2]

The same method as in Example 1 of the specification of the present application was used to manufacture a small aperture rubber latex, but (1) 0.15 parts by weight of a molecular weight controlling agent (TDDM) was added instead of 0.3 parts by weight of the same, (2) 0.02 parts by weight of TDDM (tertiary dodecyl mercaptan) was added again instead of 0.05 parts by weight of the same, and (3) the reaction temperature was increased from 65 °C to 68 °C. A large aperture rubber latex was manufactured by use of the obtained small aperture rubber latex, through the same method as that of Example 1. Graft copolymerization material was manufactured using the same method as that of Example 1.

Table 1 below shows the comparison between the graft copolymerization material manufactured by Example 1 and the graft copolymerization materials manufactured by Reference Examples 1 and 2.

[Table 1]

|   | Reference<br>Ex. 1 | Example 1 | Reference<br>Ex. 2 | Comp. Example 4<br>(only small aperture latex) |
|---|--------------------|-----------|--------------------|--|
| Large and Small Aperture<br>Rubber Latex Gel Content  | 65%                | 90%       | 98%                | 90%  |
| Large and Small Aperture<br>Rubber Latex Swelling Index                                       | 35                 | 17-18     | 8                  | 18   |
| Solidification During Graft<br>Copolymerization Reaction<br>(%) <sup>1)</sup>                 | 0.2%               | 0.12%     | 0.08%              | 0.10%  |
| Transparency of Graft<br>Copolymerization Material<br>(haze value) <sup>2)</sup> ASTM, D-1003 | 4.9                | 3.5       | 2.8                | 2.4  |
| Notch Isod Shock Strength<br>(ASTM, D-256)  | 17.5               | 16.0      | 12.0               | 3  |

1) Stability of a latex is decreased with increases in solidification.

2) Transparency is decreased with increases in haze value.

As shown in Table 1 above, if the gel content of rubber latex is high and the swelling index is low, stability of a latex and transparency become better, but shock strength becomes lower. This means that the gel content and the swelling index of rubber latex should be adjusted in order to provide a thermoplastic transparent resin, which has excellent transparency, latex stability, and shock strength.

Table 2 below shows the comparison between the rubber components of the thermoplastic transparent resin of Cited Reference 1 (U.S. Patent No. 4,767,833) and those of the present invention.

[Table 2]

|                      | Used Rubber Component |                | Average Particle<br>Diameter ( $\mu$ m) | Gel Content<br>(by weight %) | Swelling<br>Index <sup>o</sup> |
|----------------------|-----------------------|----------------|---|------------------------------|--------------------------------|
| Cited<br>Reference 1 | SBR                   | Small Aperture | 0.05-0.2                                | 30-100                       | -                              |
|                      |                       | Large Aperture | 0.2-3                                   | 10-80                        | -                              |
| Present<br>Invention | BR                    | Small Aperture | 0.06-0.15                               | 70-95                        | 12-30                          |

|  |                |          |       |       |
|--|----------------|----------|-------|-------|
|  | Large Aperture | 0.26-0.5 | 70-95 | 12-30 |
|--|----------------|----------|-------|-------|

As shown in Table 2 above, Cited Reference 1 discloses a small aperture SBR (styrene-butadiene-rubber) latex having the gel content of 30-100% and a large aperture latex having the gel content of 10-80%. Thus, the range of the gel contents disclosed in Cited Reference 1 are broad. Further, Cited Reference 1 sets the entire range of gel contents, which are utilizable in manufacturing common rubber. In addition, as shown in Table 1 of Cited Reference 1, (1) the examples of Cited Reference 1 used the gel contents of the large aperture SBR latex as being 20, 60 and 70; and (2) the comparative examples of Cited Reference 1 used the gel contents of the large aperture SBR latex as being 2, 5, 20 and 90. That is, the examples and comparative examples in Cited Reference 1 performed tests throughout the entire range of gel contents.

Unlike Cited Reference 1, (1) the present invention set an identical range of gel contents between the large and small aperture BR (butadiene-rubber) latexes; (2) the range of gel contents is narrow as 70-97%; and (3) the gel contents are set as higher values.

In general, the gel content of rubber is proportional with the degree of the cross-linking, while the swelling index of rubber is inversely proportional with the degree of the cross-linking. If the gel content of rubber latex is high and the swelling index of rubber latex is low, it is difficult for grafted monomers (e.g., methacrylic acid alkylester compounds or acrylic acid alkylester compounds, aromatic vinyl compounds, and vinylician compounds) to be diffused into a rubber latex forming a core. As a result, (1) the number of monomers, which can be swelled in the rubber latex, are small, and (2) grafted monomers are concentrated in a shell portion. In general, in order to obtain transparency, a refraction index of a core, in which a major component is a rubber latex (BR), should be identical to that of a shell, which consists of a mixture of monomers grafted in the rubber latex. As long as grafted monomers are concentrated in the shell portion, better transparency can be obtained because the refraction index may be easily adjusted.

On the basis of this fact, the present invention used a rubber latex having the high gel content of 70-97%. The present invention also increases the shock strength of the rubber latex through the use of a large aperture rubber latex having a relatively large particle diameter.

This fact is supported by Example 1 using a small and large aperture rubber latex, and Example 4 using only a small aperture rubber latex, as shown in Table 3 of the specification of the present application and Table 2 above (Examples 1 and 4 describe a rubber latex having an identical gel content and swelling index). When comparing Examples 1 and 4, Example 1 shows the 16 Notch Isod shock strength of graft copolymerization material, while Example 4 shows the 3

broad

vs,

narrow

 $GL \propto X_{LMM}$  $SI \propto \frac{1}{X_L}$

Notch Isod shock strength of graft copolymerization material. The shock strength in Example 1, which uses both the small and large aperture rubber latex, is higher than that in Example 4.

Cited Reference 1 neither suggests nor teaches the technical aspects of the present invention of (1) using a small aperture latex and defining a high gel content in order to obtain excellent transparency, and of (2) using a large aperture rubber latex in order to compensate shock strength.

In sum, since the present invention limited a gel content to 70-97% in consideration of the transparency of a thermoplastic transparent resin, the technical aspect of the present invention is entirely different from the gel content range of Cited Reference 1. Moreover, Cited Reference 1 does not mention a swelling index. Further, the present invention uses BR as a rubber latex, whereas Cited Reference 1 uses SBR as a rubber latex. Thus, the two inventions are different in terms of technical constitution. Therefore, since the present invention has technical difficulty over Cited Reference 1, the present invention has an inventive step.

That the undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine, imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patents issuing thereon;

Further declarant saith not.

Date:

*Changlong Lee*